

Behaviour of lead in uranium ore deposit; application of isotope analysis by secondary ion mass spectrometry (SIMS)

H. Isobe	Department of Environmental Safety Research, Japan Atomic Energy Research Institute, Tokai, Ibaraki, 319-1195, Japan
H. Hidaka	Department of Earth and Planetary System Sciences, Hiroshima University, Higashi-Hiroshima 739, Japan
T. Ohnuki	Department of Environmental Safety Research, Japan Atomic Energy Research Institute, Tokai, Ibaraki, 319-1195, Japan

Lead is a multi-isotope element, and each isotope has different primeval and/or radiogenic origin. The isotope composition of lead in rocks and minerals reflects their age and the migration behaviour of lead. Especially, in the vicinity of uranium deposits, lead isotope composition of minerals can provide us detailed information on the migration behaviour of uranium and lead quantitatively. When a geological event that migrates lead from a uranium ore body to surrounding formation occurs, radiogenic lead produced in the ore body commonly forms lead minerals like galena. The assemblage of uraninite and galena often occurs in uranium ore deposits. Since galena close to uraninite consists almost only radiogenic lead produced in uraninite, the formation age cannot be estimated by the Pb-Pb dating method.

Secondary Ion Mass Spectrometer (SIMS) is a powerful instrument for analysing isotope compositions of geological materials in micronmeter scale. Appropriate combination of isotope analysis by SIMS with mineralogical observation including chemical analysis by EPMA can reveal not only chronological information but also migration behaviour of lead in a uranium ore body. In this study, lead isotope analysis of the samples from the Koongarra uranium deposit, Australia was carried out by SIMS. The main ore body of the Koongarra deposit occurs in a quartz chlorite schist (Snelling, 1980). We discuss the ages of the original uraninite and the event that formed current uraninite and released lead from the ore body. The fraction of lead release is also estimated.

Experimental

SIMS analyses were carried out using a Sensitive High Resolution Ion Micro Probe (SHRIMP II) at Hiroshima University. The primary beam is O_2^- and the sputtering spot was 20 μm in diameter. Samples

were Au coated petrologic thin sections from the drill cores of the Koongarra deposit. Analysed minerals were uraninite in the primary ore body (DDH58-129) and sulphide minerals including galena (PbS), pyrite (FeS_2) and chalcopyrite (CuFeS_2) in a sample just above the primary ore body (DDH58-126.6). Pyrite grains from a barren rock (DDH60-125) at the border of alteration halo 30m in distance from DDH58-129 and 126.6 are also analysed to establish the primeval lead fraction at Koongarra.

Results and discussion

Lead isotope compositions analysed by SIMS are listed in Table 1. The abundance of ^{204}Pb indicates the degree of the primeval lead influence on the formation of minerals. The $^{204}\text{Pb}/^{206}\text{Pb}$ ratios of the Koongarra uraninite are less than 1/1000 of that of the primeval lead. Consequently, no correction was made to the analysed $^{207}\text{Pb}/^{206}\text{Pb}$ ratio for uraninite.

TABLE 1. Lead isotope ratios of minerals. n represents number of analysis.

	n	$^{207}\text{Pb}/^{206}\text{Pb}$	
		Average	σ
uraninite ¹	9	0.0767	0.0028
galena ²	9	0.1102	0.0024
pyrite ³	4	0.9594	0.0520
pyrite ³	1	0.5723	0.0018 ⁴
pyrite ³	1	0.2298	0.0011 ⁴
radiogenic lead		0.1067	—

¹ DDH58-129, ² DDH58-126.6, ³ DDH60-125, ⁴ σ represents counting error. Radiogenic lead fraction is calculated by linear regression of sulphide minerals

TABLE 2. Simultaneous equations used in the calculation. λ_1 and λ_2 are decay constants of ^{238}U and ^{235}U , W_{206} , W_{207} , W_{238} and W_{235} are atomic weight of ^{206}Pb , ^{207}Pb , ^{238}U and ^{235}U , and $^{235}\text{U}_0$ and $^{238}\text{U}_0$ are current abundance of uranium isotopes, respectively

$$\begin{aligned} (^{207}\text{Pb}/^{206}\text{Pb})_{\text{galena}} &= (1/137.88)(e^{\lambda_1 t_1} - e^{\lambda_2 t_2}) / (e^{\lambda_1 t_1} - e^{\lambda_2 t_2}) \\ (^{207}\text{Pb}/^{206}\text{Pb})_{\text{u}} &= (1/137.88)(r(e^{\lambda_1 t_1} - e^{\lambda_2 t_2}) + (e^{\lambda_2 t_2} - 1)) / (r(e^{\lambda_1 t_1} - e^{\lambda_2 t_2}) + (e^{\lambda_2 t_2} - 1)) \\ (\text{Pb}_w/\text{U}_w)_{\text{u}} &= (^{235}\text{U}_0(r(e^{\lambda_1 t_1} - e^{\lambda_2 t_2}) + (e^{\lambda_2 t_2} - 1))W_{207} + ^{238}\text{U}_0(r(e^{\lambda_1 t_1} - e^{\lambda_2 t_2}) + (e^{\lambda_2 t_2} - 1))W_{206}) / (^{235}\text{U}_0W_{235} + ^{238}\text{U}_0W_{238}) \end{aligned}$$

Sulphide minerals have various lead isotope composition. Galena close to the primary ore body has homogeneous isotope composition, and more than 99% of lead is radiogenic origin from uranium. On the other hand, pyrite from DDH60-125 has lead isotope composition close to the average terrestrial lead except two grains. These two grains have mixed composition of radiogenic and premeval fractions.

Uranium minerals in the Koongarra primary ore body including uraninite and uranyl minerals have homogeneous lead isotope (Isobe *et al.*, 1998). This suggests that lead is not released from the primary ore region since the current uraninite crystallized. Radiogenic lead in sulphide minerals surrounding the primary ore body should be released from the ore body by the event that formed the current uraninite crystals.

The ages of the original uraninite formation (t_1) and current uraninite (t_2), and the fraction of lead retained in the ore body (r) can be calculated from the lead isotope compositions of uraninite $(^{207}\text{Pb}/^{206}\text{Pb})_{\text{u}}$ and radiogenic fraction of galena $(^{207}\text{Pb}/^{206}\text{Pb})_{\text{galena}}$, and weight ratio of lead to uranium in the current uraninite $(\text{Pb}_w/\text{U}_w)_{\text{u}}$. These values have relations expressed in the simultaneous equations in Table 2. These equations can be solved numerically on t_1 , t_2 and r .

We applied this method to date uraninite and galena from the Koongarra deposit with isotope data analysed by SIMS and chemical composition of uraninite reported by Isobe *et al.* (1992). Solutions of the simultaneous equations in Table 2 for analysed results listed in Table 1 are $t_1 = 1400 \pm 60$ Ma, $t_2 = 560 \pm 70$ Ma and $r = 0.36 \pm 0.04$. Error range is estimated from standard deviations of analytical results.

The mineralization age of the Koongarra primary ore body was estimated to be less than 1650 Ma (Mass, 1989). Hills and Richards (1976) reported U-Pb dating of uraninite from Koongarra. One of five uraninite grains is plotted on the concordia curve at the age of 870 Ma, but rests of them are scattered below the concordia and do not form any definite

discordia lines. Their results indicate that U-Pb systematics of uraninite is disturbed for U-Pb method, which needs multiple grains variously, affected by the single event.

The age of 1400 ± 60 Ma calculated by the present study may show closure date of lead from the primary ore body after the formation of the deposit. Estimated formation of galena and secondary uraninite at 560 ± 70 Ma may be the age of the event which migrated lead from the primary ore body to the host rock over approximately 1m. Any apparent geological activities correspond to these ages are not reported in the region. However, the chemical and isotope compositions of the current uraninite and galena in the primary ore body suggest that the Koongarra deposit has experienced incomplete lead release from uraninite.

We conclude that the equations used here based on the simple assumption with isotope analysis of lead in uraninite and galena and chemical analysis of uraninite can be solved to estimate the ages of uraninite and galena and degree of lead migration. This method is easy to apply to uranium deposits where conventional dating methods have difficulties because of high uranium and radiogenic lead content. The information on the ages of uraninite and galena, and especially degree of lead release is essential to understand geological events concerning migration of elements in uranium ore deposits.

References

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